Nodeless superconductivity in $K_xFe_{2-y}Se_2$ single crystals revealed by low temperature specific heat

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Low temperature specific heat (SH) has been measured in $K_xFe_{2-y}Se_2$ single crystals with T_c = 32 K. The SH anomaly associated with the superconducting transition is moderate and sharp yielding a value of $\Delta C/T\mid_{T_c} = 11.6 \pm 1.0\ mJ/molK^2$. The residual SH coefficient $\gamma(0)$ in the superconducting state at T$\rightarrow$ 0 is very small with a value of about 0.39 $mJ/molK^2$. The magnetic field induced enhancement of the low-T SH exhibits a rough linear feature indicating a nodeless gap. This is further supported by the scaling based on the s-wave approach of the low-T data at different magnetic fields. A rough estimate tells that the normal state SH coefficient γ_n is about 6 $\pm$ 0.5 $mJ/molK^2$ leading to $\Delta C/\gamma_n T\mid_{T_c} = 1.93$ and placing this new superconductor in the strong coupling region.

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The discovery of high temperature superconductivity in iron pnictides has opened a new era towards the investigation on the novel superconducting mechanism.[1] One of the key issues here is about the superconducting pairing mechanism. Experimentally it was found that the superconductivity is at the vicinity of a long range antiferromagnetic (AF) order[2], the superconducting transition temperature is getting higher when this AF order is suppressed. It was also further proved that the AF spin fluctuation[3] and the multi-band effect[4] are two key factors for driving the system into superconductive. Theoretically several different pairing symmetries are anticipated. It was suggested that the pairing may be established via inter-pocket scattering of electrons between the hole pockets (around Γ point) and electron pockets (around M point), leading to the pairing manner of an isotropic gap on each pocket but with opposite signs between them (the so-called $S^{\pm}$).[5–8] The pairing picture based on the super-exchange of local moment was also proposed, which in principle could also lead to the $S^{\pm}$[9, 10], leaving the others (d-wave or full-gapped S^{++}) as perspectives with low possibility. However, the S^{++} pairing manner is specially winning the vote when the orbital fluctuation plays the important role, as argued by Kotani et al.[11]. Clearly multi-orbits, or the naturally formed multi-pockets are highly desirable for the superconductivity of all these pairing models.

Recently a new Fe-based superconducting system $A_xFe_{2-y}Se_2$ (A= alkaline metals, x$\leq$1, y$\leq$ 0.5) were discovered with the transition temperature above 30 K.[12] The interests to this fascinating system are immediate because of the two major reasons:(1) Both the band structure calculations[13, 14] and the preliminary angle resolved photo-emission spectrum (ARPES) measurements[15–17] indicate that the band near the Γ-point seems diving far below the Fermi energy, leading to the absence of the hole pockets which are widely expected for the FeAs-based systems. A consequence of these results is to question the importance of the inter hole-electron pocket scattering for the superconducting pairing. (2) The superconducting state seems occurring via a transmutation from an insulating ordered state of Fe-vacancies.[18] The question raised here is whether this insulating state originates from the Mottness, like in the cuprate,[20] or the band gap due to the reconstruction of the electronic structure when the Fe vacancies are present. Many different kind of pairing symmetries are proposed, such as nodeless d-wave,[21] S^{++} or $S^{\pm}$, all are satisfying with the basic structures. The experimental evidences about the superconducting gaps so far are quite rare. The ARPES measurements indicate isotropic gaps on the four electron pockets with a rather large gap value ($\sim$ 8-15 meV). The conclusions drawn from NMR measurements seem controversial.[22, 23] In this paper, we present the first set of data of low temperature specific heat (SH) measurements. Our detailed analysis indicates a nodeless gap with a strong coupling strength in this new superconducting system.

The $K_xFe_{2-y}Se_2$ single crystals were synthesized by the flux-growth method[19]. The typical dimension of the samples for specific heat measurements was 2$\times$2$\times$0.5 mm^3. The SH measurements were done with the thermal relaxation method on the Quantum Design instrument physical property measurement system (PPMS) with the temperature down to 2 K and magnetic field up to 9 T. The magnetic field effect on the bare SH measuring chip (including the four thermal conducting wires) of PPMS from Quantum Design was calibrated prior to the measurements on the samples, in order to remove the pseudomorphism. This becomes very essential since the contribution of the electronic SH is quite small compared to the phonon part in this particular system. The dc magneti-

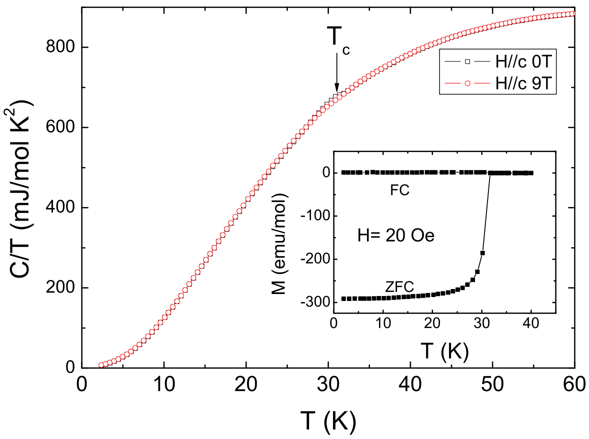

FIG. 1: (color online) Raw data of the temperature dependence of specific heat for the $K_xFe_{2-y}Se_2$ at 0T and 9T. The inset shows the temperature dependence of the dc magnetization for the same sample measured under a magnetic field of 20 Oe.

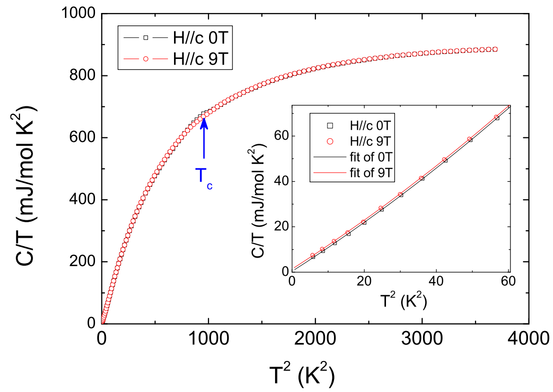

FIG. 2: (color online) The main panel is the SH data plotted as C/T vs T^2. The inset is the enlarged view of the data together with the fit (see text) in low temperature region, and no Schottky anomaly was detected.

zation measurements were done with a superconducting quantum interference device (Quantum Design, SQUID).

In Fig.1, we show the temperature dependence of specific heat and dc magnetization for the sample. The sharp transition in the magnetization and the large magnetic screening signal indicate the good quality of the sample. In the SH data at zero field, one can see a small feature of heat capacity anomaly at T_c. We will show that this SH anomaly is actually rather sharp with a reasonable magnitude.

The SH data for the sample in low temperature region are plotted as C/T vs T^2 in the inset of Fig.2. No Schottky anomaly was detected. The data below about 8 K can be fitted by

$$C(T,H) = \gamma(H)T + \beta T^3 + \eta T^5, \qquad (1)$$

where $\gamma(H)T$ is the residual SH coefficient in the magnetic field H, $\beta T^3 + \eta T^5$ is the phonon part of heat capacity. Normally it is unnecessary to consider the last term ηT^5 in the low temperature region, while this seems not the case for the present sample. One can see a slight upturn curvature in the low-T SH data C/T vs. T^2. This is understood because of the relatively low Debye temperature of the sample, as discussed below.

For the heat capacity of the sample, the phonon contribution should be identical in zero field and in magnetic field. Hence, the parameters β and η should be the same for 0T and 9T. This is a constraint on the fitting process of the data. By fitting the SH data in 0T and 9T using Eq.(1), we obtained $\gamma(0) \approx 0.394$ mJ/mol K^2, $\gamma(9T) \approx 1.4$ mJ/mol K^2, $\beta \approx 1.018$ mJ/mol K^4 and $\eta \approx 0.003$ mJ/mol K^6. Using the obtained value of β and the relation $\Theta_D = (12\pi^4 k_B N_A Z/5\beta)^{1/3}$, where $N_A = 6.02 \times 10^{23}$ mol^{-1} is the Avogadro constant, Z = 5 is the number of atoms in one unit cell, we get the Debye temperature $\Theta_D \approx 212$ K, which is relatively small, compared to other FeAs-based superconductors[24, 25].

Fig.3(a) shows the enlarged view of the SH data near the transition temperature plotted as C/T vs T. One can see a clear SH anomaly at T_c in 0T, and when a magnetic field is applied, the SH anomaly was weakened and shifted to lower temperatures. Fig.3(b) shows the difference of the SH data between 0T and 9T. The SH anomaly at T_c is more obvious. We can evaluate the height of the SH anomaly $\Delta C/T|_{Tc}$ near T_c from the difference of C/T at 0T and 9T. The estimated anomaly $\Delta C/T|_{Tc}$ is about 11.6 $\pm$ 1 $mJ/molK^2$. For the optimally doped (Ba,K)Fe_2As_2 and Ba(Fe,Co)$_2As_2$, the SH anomaly $\Delta C/T|_{Tc}$ are about 98 $mJ/molK^2$ and 28.6 $mJ/molK^2$, respectively[25–29]. The SH anomaly for $K_xFe_{2-y}Se_2$ is also smaller than other FeAs-based superconductors. The SH anomaly looks rather sharp (it starts at about 32.9 K and ends at 30.9 K). We must emphasize that to use the data measured at 9 T as the background to deduce the SH anomaly is reasonable. As we address below that a magnetic field of 9 T should have lowered down the transition temperature of about 5-6 K (with the upper critical field $H^c_{c2(0)} \approx 48T$), being much larger than the width of the SH anomaly. The rather sharp SH anomaly is very different from that in the underdoped cuprates in which a long-tail of electronic SH was observed far into the normal state.[30] This was interpreted as the fluctuating superconductivity. This may suggest that, although having a low superfluid density and relatively higher anisotropy,[31] the superconducting transition in $A_xFe_{2-y}Se_2$ superconductors can still be described quite well by the critical mean field theory without the necessity of categorizing it into the strong critical fluctuation.

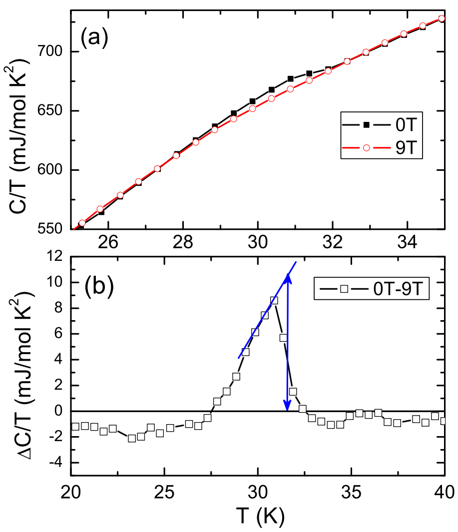

FIG. 3: (color online) The upper panel is the enlarged view of the SH data near the transition temperature plotted as C/T vs T. The lower panel shows the difference of the SH data between 0T and 9T. The blue solid lines here are just guides to the eyes for showing how we determine the height of the SH anomaly.

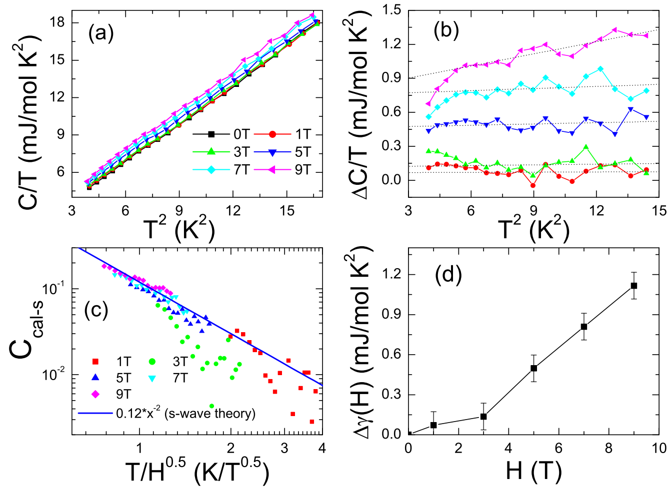

FIG. 4: (color online) (a) The SH data in low temperature region under various magnetic fields. (b) The difference of the SH data between a particular magnetic field and zero field. The dashed lines are the linear fit of this difference between 2.4 K - 4 K. The slight bending down of the data at 5, 7, 9 T was recognized as due to the unsuccessful removing of the magnetic field effect on the SH measuring chip. (c) Scaling of the data according to the s-wave scenario (symbols) $C_{cal-s} = [C(H) - C(0)]/T^3$ vs $T/\sqrt{H}$, the dashed line represents the theoretical expression. (d) Field dependence of the field-induced term $\Delta\gamma(H) = [C(T,H) - C(T,0)]/T$ at T = 3K.

Fig.4(a) shows the SH data plotted as C/T vs T^2 in low temperature region under various magnetic fields. One can see that the magnetic field enhances the SH coefficient progressively, indicating the generating the quasiparticle density of sates. Fig.4(b) presents the difference of the SH data between a typical magnetic field and zero field and the dashed lines are the linear fit of the difference in the temperature region of 2.4 K to 4 K. The slight bending down of the data here below about 2.4 K at fields of 5, 7, 9 T was recognized as due to the unsuccessful removing of the magnetic field effect on the SH measuring chip. And this would have a more obvious effect on high magnetic field data, and especially the low-T region. As shown in Fig.4(b), the difference of SH data for 9T and 0T is a little tilted , rather than a rough constant, so we extracted the data at T = 3K as the field induced term of SH. Using the linear fit (between 2.4 and 4 K) in panel (b), we can obtain the magnetic field induced enhancement of the low-T SH, and the extracted data at T = 3K are shown in Fig.4(d). It is clear that the field induced term exhibits a roughly linear field dependence. In combination with the fact that a very small residual SH term was observed in the superconducting state at T→0, we tempt to conclude a nodeless gap.

In order to further confirm this point, we analyzed the SH data in finite-temperature region in the mixed state. It is known that the quasiparticle excitations in superconductors with different gap symmetries can be obviously distinct. In s-wave superconductors, the inner-core states dominate the quasiparticle excitations, and consequently a simple scaling law $C_{QP}/T^3 \approx C_{core}/T^3 = (\gamma_n/H_{c2(0)} \times (T/\sqrt{H})^{-2}$ for the fully gapped superconductors is expected, where C_{QP} and C_{core} are the specific heat of the quasiparticles induced by field and that from the vortex cores in the mixed state, respectively. The scaling result of the field-induced term in the mixed state with the s-wave condition is presented in Fig.4(c). One can see that all the data at different magnetic fields can be roughly scaled to the straight blue line, which reflects the theoretical curve $C_{cal-s} = 0.12(T/\sqrt{H})^{-2}$. Generally, this prefactor $\gamma_n/H_{c2(0)} = 0.12mJ/(molK^2T)$ is consistent with the magnitude of the slope of the line in Fig.4(d). Using the value of $H^c_{c2(0)} \approx 48T$[19], we estimate the value of normal-state electron SH coefficient γ_n to be 5.8 $mJ/molK^2$, which is a small value compared to other FeAs-based superconductors[25–29].

As far as we know, reliable calculated values for the normal state DOS and thus the SH coefficient of this new superconductor are still lacking because of the uncertainties of the structures of these Fe-vacancies. The $\gamma_n \approx 6$ $mJ/molK^2$ found here will give some hint on the band structures as well as understanding the ARPES data. The value $\Delta C/\gamma_n T\mid_{T_c} = 1.93$ clearly places the system in the strong coupling camp, since the weak coupling BCS theory gives 1.43. Furthermore, the very small residual SH coefficient $\gamma(0) \approx 0.39$ $mJ/molK^2$ together with the

s-wave scaling excludes the nodal gaps in this system. This is consistent with the ARPES data so far, and in contradicting with the NMR data.[23] The small $\gamma(0)$ detected here excludes also the chemical phase separation picture, since otherwise a much significant value, arising from the non-superconducting normal metallic regions, should be observed. However, if the system is chemically separated into the superconducting regions and the insulating regions which are fully gapped, this is of course acceptable. Our results here is also against with the nodeless d-wave picture since that kind of pairing is certainly suffered sensitively from the impurity scattering which would give rise to a large quasiparticle density of states detectable by specific heat. The sharp SH anomaly found here indicates that the present system does not have a strong critical fluctuation which appears in the underdoped cuprates.

In summary, we measured the low temperature SH of single crystal $K_xFe_{2-y}Se_2$ in various magnetic fields. The SH anomaly is observed at $T_c = 32$K, and the height $\Delta C/T\mid_{T_c}$ is about 11.6 $mJ/molK^2$. From the low temperature part of the SH data, we obtained the field induced enhancement of the low-T SH, which exhibits a roughly linear field dependence, indicating a nodeless gap. We also analyzed the data with the s-wave scaling law, and found that the data roughly obey this law, indicating again an s-wave gap. These two approaches are self-consistent each other. The Debye temperature and the normal-state electronic SH coefficient were also estimated, both are smaller than that in other FeAs-based superconductors.

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